

Spin dynamics in a hole-doped $S = 1/2$ Heisenberg antiferromagnet with a disordered ground state

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Only 3% hole doping by Li is sufficient to suppress the long-range antiferromagnetic order in La_2CuO_4 . Spin dynamics in such a disordered state was investigated with measurements of the dynamic magnetic structure factor $S(\omega, \mathbf{q})$, using cold neutron spectroscopy, for $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$. The $S(\omega, \mathbf{q})$ is found to sharply peak at (π, π) , and its dynamics to be relaxational. Confirming theoretical expectation for the quantum disordered 2D $S = 1/2$ Heisenberg antiferromagnet, the energy scale saturates at a finite value at low temperatures. Possible connection to the “pseudo spin gap” phenomenon observed in the NMR/NQR studies on underdoped cuprates is discussed.

Stimulated by the discovery of cuprate superconductors, which derive from doping charge carriers into weakly coupled two-dimensional (2D) antiferromagnetic CuO_2 planes, there has been great interest in 2D $S=1/2$ Heisenberg antiferromagnet (HAF) with dominant nearest-neighbor exchange interactions on a square lattice. It is now generally accepted that there is long-range Néel order at $T = 0$ for such a system[1]. Finite-temperature magnetic properties[2] are in good agreement with theoretical predictions[3].

The situation more closely related to superconductivity in cuprates, namely, 2D $S=1/2$ HAF with doped holes, however, is much less understood. The parent compounds such as La_2CuO_4 are charge-transfer materials[4], therefore, the doped hole is a charge with $S = 1/2$ located at an O site. However, the formation of the Zhang-Rice singlet[5] allows an effective description of the hole as a spinless charge at a Cu site. Strong suppression of the Néel order by holes can be accounted for by long-range topological disturbances accompanying holes[6]. In addition, hole motion is also disruptive to the Néel order[7]. For hole-doped La_2CuO_4 without the Néel order, two type of incommensurate antiferromagnetic correlations have been discovered experimentally when holes introduced by dopants such as Sr or Ba are mobile[8]. When holes are loosely bound in the case of Li-doped La_2CuO_4 [9], antiferromagnetic correlations remain commensurate[10]. Microscopic understanding of interaction between holes and spins on the square lattice remains a major challenge in condensed matter research[11, 12].

An alternative approach to 2D $S = 1/2$ HAF with doped holes is based on quantum phase transition. Building on the success of the quantum non-linear σ model as an effective low temperature theory for 2D $S=1/2$ HAF[3, 13], effect of doping is simulated by a frustration parameter g , which at a critical value g_c suppresses the long-range Néel order at $T = 0$ [3, 14]. The advantage of this approach is that aspects of spin dynamics can be predicted from general theoretical arguments for dynamic critical phenomena before microscopic theory is

established. The magnetically disordered state in the g - T plane is divided into three physical regimes: the renormalized classic, quantum critical (QC) and quantum disordered (QD). For doped cuprates with disordered ground state ($g > g_c$), only the QC and QD regimes are relevant. Quantum critical theory[14, 15] predicts that the energy scale, Γ , is proportional to T in the QC regime at higher T . Inelastic neutron scattering studies on Ba or Sr doped La_2CuO_4 have confirmed this prediction over an extraordinarily wide energy ($\hbar\omega \leq 90$ meV) and temperature ($T \leq 500$ K) range[16, 17]. At lower T in the QD regime, Γ is expected theoretically to saturate at a finite value, Γ_0 . Here we report a direct observation of such a behavior in a cold neutron inelastic neutron scattering study on $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$. With an energy resolution of 0.1 meV in term of the full-width-at-half-maximum of incoherent scattering, we focused on low energy spin dynamics, relevant in search for Γ_0 . Possible connection of the QD spin dynamics to the so-called “pseudo spin gap” phenomenon observed in the NMR/NQR studies on hole-doped La_2CuO_4 is discussed.

A single crystal of $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$, weighing 2.1 g, was grown in CuO flux, using isotopically enriched ^7Li (98.4%) to reduce neutron absorption. The crystal has orthorhombic $Cmca$ symmetry with lattice parameters $a = 5.351\text{\AA}$, $b = 13.15\text{\AA}$ and $c = 5.386\text{\AA}$ at 295 K. Using this orthorhombic unit cell to label reciprocal \mathbf{q} space, the (π, π) point in the square lattice notation splits to (100) and (001) points. Neutron scattering signal is observed at (100) type but not at (001) type Bragg points, as in the stoichiometric antiferromagnet La_2CuO_4 [18]. Measurements of seven independent (100) type of Bragg peaks, using thermal neutrons to reach 6\AA^{-1} , confirm the magnetic origin of these peaks. In the remainder of this study, we will focus on spin dynamics near $\mathbf{Q}=(100)$, using cold neutron triple-axis spectrometer SPINS at NIST with fixed $E_f = 3.7$ meV or $E_f = 5$ meV. The (002) reflection of pyrolytic graphite was used for both the monochromator and analyzer. A cold Be or BeO filter was used to eliminate higher order neutrons. Horizontal Soller slits of $80'$ were used before and after the sample. Temperature

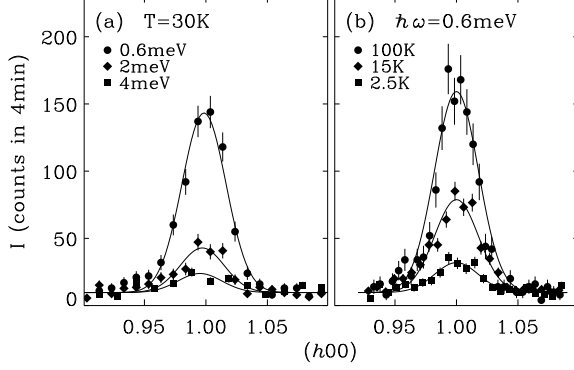


FIG. 1: (a) Const.- $\hbar\omega$ scans with various energy transfers at 30 K. (b) Constant- $\hbar\omega = 0.6\text{meV}$ scan at various temperatures. The $\mathbf{q}=(100)$ here in the orthorhombic notation corresponds to the (π, π) point of the CuO_2 square plane.

of the sample was regulated by a pumped He cryostat.

Fig. 1 shows some constant-energy ($\hbar\omega$) scans around $\mathbf{Q}=(100)$ at various energies and temperatures, which roughly cover the energy and temperature range of this study. All the peaks at (π, π) are resolution-limited. This means that dynamic spin clusters with nearest-neighbor antiferromagnetic alignment in the CuO_2 plane have grown to substantial size. From the half-width-at-half-maximum of the peaks, a *lower bound* of magnetic correlation length can be estimated, namely, $\xi > 42\text{\AA}$ below ~ 100 K. The correlation length is much longer than the mean distance, 15\AA , between Li dopants. Therefore, consistent with expectation from microscopic theory by Haas et al.[6], antiferromagnetic correlations in our sample are not simply impurity limited.

Dynamics of the antiferromagnetically correlated spin clusters in $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$ are probed by energy scans at $\mathbf{Q}=(100)$ at various temperatures (Fig. 2). Since magnetic intensity is confined within resolution at (100) , the energy scan at $(1.39, 0, 0)$, which is T and $\hbar\omega$ -independent (refer to triangles in Fig. 2), offers a good measure of background. Lowering temperature from 150 K, magnetic intensity at lower energies increases at the expense of magnetic intensity at higher energies, as shown by filled and open diamonds. This is typical for paramagnetic fluctuations, as magnetic susceptibility increases while fluctuation energy decreases with lowering temperature[19]. Upon further lowering temperature, however, magnetic intensity below $\sim 1.5\text{meV}$ is progressively suppressed. At 1.5 K, a peak at a finite energy can be clearly discerned. We would like to emphasize here that there is no real gap in the spin excitation spectra in Fig. 2. As will be shown later, spectra in Fig. 2 can be well described by relaxational spin dynamics. Theoretically, both gapped and gapless spin dynamics have been found in different toy models in the QD regime[14].

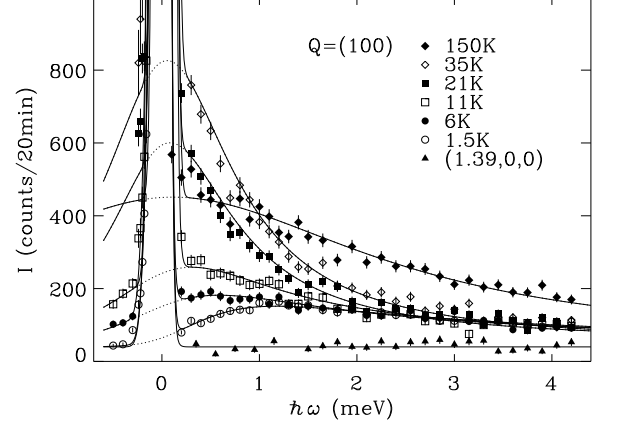


FIG. 2: Const.- $\mathbf{Q}=(100)$ scans from 1.5 to 150 K. The solid curves at finite $\hbar\omega$ and their dot-line continuation near $\omega = 0$ are theoretical fit to Eqs. (2) and (3) with parameters Γ and $\chi_{\mathbf{Q}}$ shown in Fig. 3. Triangles represent a measured flat background at $(1.39, 0, 0)$.

The spectra in Fig. 2 also differ from inelastic neutron scattering spectra from conventional, heavy fermion, and cuprate superconductors, which do show spin or pseudo spin gap due to superconducting transition[20, 21, 22].

Magnetic neutron scattering intensity is a convolution of instrument resolution function with dynamic magnetic structure factor $S(\omega, \mathbf{q})$. Since \mathbf{q} scans are resolution-limited at $\mathbf{Q}=(\pi, \pi)$, it is convenient to write $S(\omega, \mathbf{q})$ as

$$S(\omega, \mathbf{q}) = \frac{1}{\pi} \frac{1}{1 - \exp(-\hbar\omega/k_B T)} \chi''_{\mathbf{Q}}(\omega) F_{\omega}(\mathbf{q}), \quad (1)$$

where $\int d\mathbf{q} F_{\omega}(\mathbf{q}) = 1$ [16]. Sharp structure of $F_{\omega}(\mathbf{q})$ peaking at (π, π) can not be resolved in this work. Data in Fig. 2 thus represent *local* dynamic magnetic structure factor

$$\begin{aligned} S_{\mathbf{Q}}(\omega) &\equiv \int d\mathbf{q} S(\omega, \mathbf{q}) \\ &= \frac{1}{\pi} \frac{1}{1 - \exp(-\hbar\omega/k_B T)} \chi''_{\mathbf{Q}}(\omega), \end{aligned} \quad (2)$$

where $\chi''_{\mathbf{Q}}(\omega)$ is the imaginary part of the *local* dynamic magnetic susceptibility. Relaxational spin dynamics at low energy is generally described by[23]

$$\chi''_{\mathbf{Q}}(\omega) = \frac{\hbar\omega\chi_{\mathbf{Q}}\Gamma}{(\hbar\omega)^2 + \Gamma^2}, \quad (3)$$

where Γ is the relaxation energy and $\chi_{\mathbf{Q}}$ represents spectral intensity. For scan at each temperature in Fig. 2, Γ and $\chi_{\mathbf{Q}}$ are extracted by least squared fit of the data to Eqs (2) and (3), plus the usual elastic/incoherent peak, convoluted with instrument resolution. A flat background measured at $\mathbf{q}=(1.39, 0, 0)$ (refer to triangle) is included in the fitting. The consistency between the theoretical curves and measured data in Fig. 2 is very satisfactory.

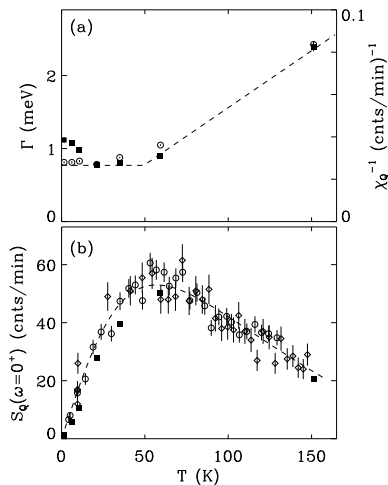


FIG. 3: (a) Measured temperature dependence of Γ (squares) and χ_Q^{-1} (circles) for $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$ [Refer to Eq. (3)]. (b) Temperature dependence of quasielastic magnetic intensity, $S_Q(\omega = 0^+)$. The filled squares are calculated using Eq. (5) from Γ and χ_Q in (a), and open circles and diamonds were measured at low energy, $\hbar\omega = 0.2$ meV, during warming and cooling, respectively.

Experimental Γ and χ_Q^{-1} as a function of temperature are shown in Fig. 3(a) by square and circle, respectively. Clearly, Γ and χ_Q^{-1} saturate at finite values at low temperatures. It is well known for conventional magnetic materials that in paramagnetic phase, Γ is a monotonically increasing function of T which is zero at magnetic transition temperature[19]. This behavior applies also to spin-freezing transition of classic spin glasses such as Cu-Mn [24]. When magnetic transition is prevented by frustration, the monotonic behavior for Γ still persists[25]. Temperature independent Γ has been observed in intermediate-valence compounds[26]. However, there is no magnetic correlations between rare-earth ions in these materials and the Γ reflects valence fluctuations. In $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$, magnetic correlations are substantial with $\xi > 42\text{\AA}$. What we observe here is consistent with theoretical expectation for 2D $S=1/2$ HAF with the QD ground state[15]: $\Gamma(T) \sim \max[k_B T, (g - g_c)^{z\nu}]$. The dashed line in Fig. 3(a), $\Gamma(T) = \max[0.18k_B T, \Gamma_0]$ with $\Gamma_0 = 0.77\text{meV}$, captures the main feature of our data.

In hole-doped La_2CuO_4 , the g is physically the doping concentration. Besides frustrating the long-range antiferromagnetic order, doping inevitably introduces disorders. The spin freezing temperature T_{sf} measured with μSR for Li-doped La_2CuO_4 [27] is nearly identical to that for Sr-doped La_2CuO_4 [28]. For $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$, $T_{sf} = 8$ K. Up to now, little has been predicted for $S(\omega, \mathbf{q})$ for realistic spin glasses. In ferromagnetic reentrant spin glasses, spin dynamics expected of a ferromagnet has been experimentally observed within a finite range of \mathbf{q} of magnetic zone center in the temper-

ature interval $T_{sf} < T < T_C$ [29]. Along this line, one would expect the QD behavior to hold between ~ 10 and ~ 60 K for $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$, refer to Fig. 3(a). In addition, below spin freezing, Γ from spin clusters at (π, π) in $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$ remains finite. There is no sign of freezing $\Gamma \rightarrow 0$, which is a universal feature for classic spin glasses. This suggests that inelastic magnetic intensity in Fig. 2 comes from spin clusters which do not participate in spin freezing. The upturn of Γ in Fig. 3(a) below 11 K may be attributed to modification to the QD dynamics due to coupling of the fluctuating spin clusters to freezing spin clusters.

One motivation of this low-energy neutron scattering work is to make connection to Cu nuclear resonance NMR/NQR results. Soon after the discovery of cuprate superconductivity, anomalous suppression of low energy spin fluctuations was discovered in *normal* state of various family of underdoped cuprates[30, 31]. Refer to Ref. [23] for a historic review of this so-called “pseudo spin-gap” phenomenon (PSG). It has been discussed variously as due to, e.g., a temperature dependent magnetic correlation length[32, 33], resonating valence bond singlet pairing[34], activation gaps of stripe glasses[35, 36], or incipient superconducting order[21]. In the La_2CuO_4 family, extensive inelastic neutron scattering studies have found no such gap[16, 17], except the spin gap due to superconducting transition[22].

The nuclear spin-lattice relaxation rate $1/T_1$ is a weighted summation of dynamic magnetic structure factor $S(\omega, \mathbf{q})$ over the Brillouin zone at small $\omega = 0^+$,

$$T_1^{-1} = \sum_{\mathbf{q}} |A(\mathbf{q})|^2 S(0^+, \mathbf{q}), \quad (4)$$

where hyperfine coupling $|A(\mathbf{q})|^2$ peaks at (π, π) for Cu NMR/NQR[23]. Using Eqs. (1)-(3) for $S(\omega, \mathbf{q})$,

$$\begin{aligned} T_1^{-1} &= S_Q(0^+) \sum_{\mathbf{q}} |A(\mathbf{q})|^2 F_\omega(\mathbf{q}) \\ &\sim S_Q(0) = \frac{\chi_Q k_B T}{\pi \Gamma}. \end{aligned} \quad (5)$$

In Fig. 3(b), $S_Q(0)$ calculated using Eq. (5) from the experimentally determined χ_Q and Γ in Fig. 3(a) are shown together with $S_Q(\omega)$ measured at $\hbar\omega = 0.2$ meV, which is the lowest energy transfer without significant elastic contamination (refer to Fig. 2). Circles (diamonds) were measured during warming (cooling) cycle. These data, $S_Q(\omega \sim 0) \sim 1/T_1$, bear remarkable resemblance to $1/T_1$ observed in Sr-doped La_2CuO_4 of similar hole concentration[31]. The reduction of $1/T_1$ at low T below the extrapolation from high T behavior is the experimental observation termed “pseudo spin gap” in NMR/NQR studies on cuprates. We have shown that there is no gap in spin excitation spectra for $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$. The apparent PSG behavior in Fig. 3(b) is merely a consequence of a saturating Γ at low T , which is expected for 2D

$S=1/2$ HAF of $g > g_c$ with the QD ground state. Thus a possible link between the NMR/NQR PSG behavior with the QD state is suggested by this study; specifically, the PSG phenomenon may not require a pseudo gap for its explanation.

From this point of view, if we take doped cuprates without the Néel order as the experimental realization of $g > g_c$, then the ubiquitous NMR/NQR PSG behavior is expected as a consequence of a finite $\Gamma_0 \sim (g - g_c)^{2\nu}$. The absence of spin or pseudo spin gap from inelastic neutron scattering studies can then also be reconciled with the PSG from NMR/NQR studies. Similar NMR/NQR spectra for underdoped La_2CuO_4 [28, 35, 36, 37] may also be understood as a consequence that the dopant elements, dopant location (in or out-of-plane) and crystal structure (orthorhombic or tetragonal) are irrelevant perturbations to the QD regime. As such, study of the PSG phenomenon should pay at least as much attention to saturating Γ as to the presence of a real spin gap.

In summary, we found dynamic spin clusters in hole-doped $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$ to remain (π, π) -correlated. Spin dynamics of such clusters, with correlation length much larger than mean impurity distance below 150 K, is relaxational at (π, π) . Confirming predictions of quantum critical theory for 2D $S = 1/2$ Heisenberg antiferromagnet in the quantum disordered regime, the energy scale Γ of $\text{La}_2\text{Cu}_{0.94}\text{Li}_{0.06}\text{O}_4$ saturates around 0.8 meV below ~ 60 K. We discuss the possible connection between the so-called “pseudo spin-gap” phenomenon discovered in NMR/NQR studies on underdoped cuprates and the “quantum disordered” behavior observed here. In other words, the NMR/NQR “pseudo spin-gap” phenomenon may be accounted for without pseudo spin gap in spin excitation spectra.

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